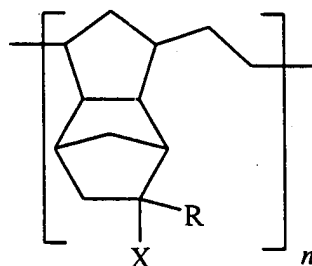## Process for the production of polyolefinic optical films

The invention relates to a process for the production of poly-
olefin films by casting a polymer solution onto a substrate.
5   In particular, it relates to a process for producing optical
films from polyolefins of the formula

in which at every occurrence of the substituents R and X they
are either both hydrogen or R is methyl and X is a polar group,
10   and n is a number from 10 to 1000, by casting solutions of the
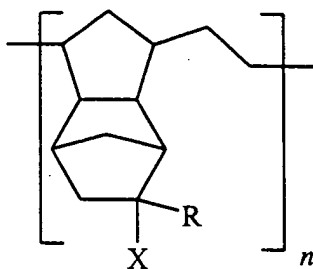polyolefins in organic solvents onto a substrate and evaporating
the solvents.

Polyolefins of the above formula are known for their good
15   optical properties and are commercially available by way of
example with the name ARTON® (X = methoxycarbonyl, producer:
Japan Synthetic Rubber Co.). A requirement for various appli-
cations is to use this material to produce optical films which
have not only constant thickness but also a smooth and defect-
20   free surface, and are free from streaking, and in particular
have minimum and constant optical retardation. The only method
of obtaining this combination of properties, if indeed it can be
obtained, is to use a casting process in which a solution of the
polymer is cast onto a substrate and solidified via evaporation
25   of the solvent, and finally in the form of a film is peeled away
from the substrate. However, it has been found that use of the
usual conditions for producing other optical films (e.g. from
cellulose triacetate) by the casting process does not give films
which meet all of the requirements.
30

It was therefore an object of the present invention to provide a
production process for optical films from the abovementioned

polyolefins which in particular gives films with extremely low optical retardation.

The invention achieves this object via the process as claimed in
5   claim 1.

It has been found that films with very low optical retardation can be produced from polyolefins of the formula

10   in which at every occurrence of R and X they are either both hydrogen or R is methyl and X is a polar group, and *n* is a number from 10 to 1000, by casting a solution of the polyolefin in an organic solvent onto a substrate and evaporating the solvent, if

15   (i)   the polyolefin is dissolved in an organic solvent or solvent mixture, and

(ii)   the solution obtained in this way is cast onto a smooth substrate in an atmosphere comprising at least 1% by volume of solvent vapor at a temperature below the boiling point of the
20   solvent, with substantially laminar gas flow being maintained over the casting substrate,

(iii)   the solvent is evaporated to obtain a self-supporting film, and

(iv)   the film is peeled away from the substrate and dried at
25   a temperature rising to 70-140 °C, without any resultant orientation of the film.

The above formula encompasses homo- and copolymers, depending on whether at each occurrence R and X are identical or different.

30

In principle, suitable solvents are any of the organic solvents of low polarity in which the polyolefin used has sufficient solubility and which have a boiling point below the glass

transition temperature of the polyolefin. Among these, by way of example, are halogenated hydrocarbons, such as dichloromethane or dichloroethane, aromatic hydrocarbons, such as toluene or xylene, aliphatic and cycloaliphatic hydrocarbons, such as
5 cyclohexane, and also mixtures of the solvents mentioned.
The solvents and solvent mixtures whose use is preferred have been selected from the groups consisting of dichloromethane, toluene, cyclohexane, and their mixtures. Particular preference is given to dichloromethane and solvent mixtures comprising
10 dichloromethane. Those used may be mixtures of dichloromethane with other volatile solvents, such as methanol, the dichloromethane advantageously being the main constituent.

The casting solutions advantageously also comprise additives,
15 such as heat stabilizers, to inhibit thermal degradation, for example during the drying process, and/or release agents, to facilitate the peeling-away of the casting substrate. Examples of suitable release agents are detergents, plasticizers, such as phosphoric, phthalic or adipic esters, metal soaps, fatty acid
20 amides, or polyethylene glycols, and derivatives of these, such as ethers or esters.

The casting substrate used preferably comprises a continuous belt, for example composed of polished stainless steel or of a
25 polymer film.
In another preferred embodiment, the casting substrate used comprises a polymer film, for example composed of Teflon or polyester. In this case, the two sub-steps "peeling away from the substrate" and drying in step (iv) of the process may, where
30 appropriate, also be carried out in reverse sequence, by drying and winding-up the casting substrate and the cast film together, and not separating the substrate from the film until a subsequent step of the process has been reached.

35 The substantially laminar gas flow is advantageously achieved by introducing solvent-containing gas (preferably air) in the vicinity of the casting gap in such a way that the gas proceeds in the same direction as the casting substrate and the velocity

of the gas relative to that of the substrate is very small or zero. In continuous operation, the solvent-containing gas is advantageously drawn off at the end of the drying zone and recycled by way of a condenser. The solvent content can thus easily be adjusted via change of the condenser temperature, as determined by the vapor pressure curve, and the condensed solvent can be reclaimed.

The casting procedure is preferably carried out with a casting solution temperature which is below the solvent boiling point by at least 2 K, particularly preferably 10-20 K. If the casting temperature is too close to the boiling point of the solvent, there is a risk that uncontrolled evaporation will form inhomogeneous regions.

The inventive process is preferably used to produce films from polyolefins of the above formula in which at least some of the substituents X are C_{1-4}-alkoxycarbonyl groups.

Particular preference is given to polyolefins in which at least some of the substituents X are methoxycarbonyl groups.

Very particular preference is given to polyolefins in which some of the substituents R and X are hydrogen and the remainder are methyl and methoxycarbonyl. These copolymers may either be block copolymers or else be random or alternating copolymers.

The concentration of the polyolefin in the casting solution is preferably from 10 to 40% by weight, particularly preferably from 20 to 35% by weight.

The thickness of the films which can be produced according to the invention is preferably from 30 to 200 μm. The inventive process can produce films with an optical retardation of less than 10 nm at 100 μm thickness and a variation in optical retardation in the longitudinal and transverse direction of only about ±1 nm (at 100 μm thickness). Very low variation in optical retardation is therefore ensured in a subsequent stretching

process to produce optically functional films (compensation and retardation films for LCD, λ/2 films, and λ/4 films, etc.).

In order to achieve a particularly low residual content of solvent (≪1%), the film can, where appropriate, be subjected to a second drying process under substantially identical temperature conditions, but advantageously with a lower web speed of the film.

The examples below illustrate the conduct of the inventive process but should not be regarded as a restriction.

Example 1
Production of a cast film from ARTON® G with a thickness of 100 µm

A homogeneous solution was prepared from 5000 kg of ARTON® G (homopolymer, R = methyl, X = methoxycarbonyl) and 13182 kg of dichloromethane, with stirring and slight heating, and was filtered in a filter press through a filter composed of a layer of calmuc fabric, a metal sieve with mesh width 12 µm, and a layer of cotton batiste. The filtered solution was heated to 40 °C for degassing and then temperature-controlled to 33 °C and cast in an atmosphere with about 3% by volume content of dichloromethane vapor and having a temperature of 34 °C, at the required thickness (casting gap about 500 µm) onto a polished circulating steel belt of length 60 m and width about 1.27 m, circulating at 3.6 m/min. The air comprising dichloromethane was introduced into the region of the casting gap in such a way as to give a linear gas velocity of about 2-5 m/s in the direction of the belt (corresponding to a relative velocity of about ±1.5 m/s). The temperature in the belt duct was raised toward the take-off point in stages to about 60 °C, and the film formed was peeled away. This was then dried over a length of about 260 m at a temperature rising in stages from about 60 °C in the region of the first 120 m of length to about 115 °C in the final third, and finally, after cooling, was cut to a width of 1010 mm, and wound up.

Once stationary-state operating conditions had been established, the film obtained had a residual solvent content of about 1.45% by weight and an optical retardation of about 10 nm.

5 **Example 2**
Production of a cast film from ARTON® G with a thickness of 100 μm

Using a method based on Example 1, a solution was prepared from
10 3840 kg of ARTON® G and 8939 kg of dichloromethane, and cast, using a belt velocity of 4.9 m/min. The resultant film with residual solvent content (determined to ASTM 1003) of 1.34% by weight, an optical retardation (measured at 632 nm) of 9 nm, and haze of 0.27% was first wound up and then after-dried in a
15 continuous dryer at a temperature of 125-132 °C over a length of 121 m at a velocity of 2 m/min. After this after-drying, the residual solvent content was 0.08% by weight, and the optical retardation was 9.5 nm. The haze had risen very slightly to 0.45%.

20

Example 3
Production of a cast film from ARTON® R with a thickness of 50 μm

25 Using a method based on Example 1, a solution was prepared from 1114 kg of ARTON® R (copolymer, R = H, methyl; X = H, methoxy-carbonyl) and 2936 kg of dichloromethane, and was cast at 23 °C (solution temperature) in an atmosphere comprising about 2.5% by volume of dichloromethane vapor at a temperature of 32 °C onto a
30 continuous steel belt of length 28 m and width about 1.20 m, circulating at 2.0 m/min. The casting gap width was about 250 μm. The air temperature for the drying in the belt duct was increased toward the take-off point in stages to 114 °C, and then the film was peeled away. The film was then heated continu-
35 ously over a length of 190 m to a maximum temperature of 95 °C, and then, after cooling, cut to a width of 1010 mm, and wound up. Once stationary-state operating conditions had been achieved, the resultant film had a thickness of 49±1 μm, resi-

dual solvent content of 0.8%, and optical retardation of 1-2 nm. The haze measured was below 0.2%.

Example 4

5 **Production of a cast film from ARTON® G**

A solution of ARTON® G (34.5% by weight) in toluene was cast with the aid of a doctor blade at a layer thickness of 400 µm onto a glass plate and dried at 120 °C. After a drying period of 10 35 min., the film could be released completely from the plate, but continued to exhibit a marked tendency to roll, which reduced greatly after 60 min.

Example 5

15 **Production of a cast film from ARTON® G**

The procedure was as described in Example 4, but the casting solution was treated with 0.01% by weight of release agent (polyethylene glycol ester), and a metal plate was used as 20 casting substrate. Good release of the film from the plate was possible after as little as 20 min. of drying at 120 °C.